IMPLEMENTATION OF ISOTOPE CORRELATION TECHNIQUE FOR SAFEGUARDS

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1. Introduction

The isotopic correlation technique (ICT) is based on the fundamental physics principle that the isotopic compositions of nuclear material in fuel cycle systems contain information regarding the design and history of nuclear material flow from fuel fabrication, reactor operation, and through input to the reprocessing plant. Isotopic Correlation in conjunction with the gravimetric (or Pu/U) method for mass determination can be developed to provide an independent in-field verification of the reprocessing input accountancy at the dissolver and/or accountancy stage of the reprocessing plant. The use of fission product and/or heavy element isotopic ratios for verifying mass input to reprocessing plants have been explored by many investigations.[1-11]

The Argonne National Laboratory program in isotope correlation techniques is based on three-dimensional reactor physics calculations of characteristic geometries/composition in each reactor class. The ANL/ICT methodology has been implemented for the analysis of the ESARDA reprocessing input verification (RIV) working group's benchmark exercise.

The ESARDA exercise to test the performance of ICT procedures has been outlined and reported by Messr. A. Giacometti.[6] The benchmark exercise was organized using reprocessing data from the La Hague plant and the reactor data from the Obrigheim power plant operation. These data consisted of mixed batches of Obrigheim assemblies, total and partial assemblies with various initial enrichments, reprocessed at the COGEMA facility. The data were divided into two groups: Set A, containing 24 batches, served as the reference data; Set B, containing 29 batches, included intentional anomalies in a fraction of the batches. Set B was to test the ability of the methodology to ascertain anomalies in the reprocessing data. The overall results of the exercise is being planned by the RIV working group for a future presentation.

This paper presents only the DOE/ANL study effort in developing the isotopic correlation technique analysis and diagnostic algorithms as a contribution to the ESARDA benchmark exercise.

2. ESARDA Single-Correlation Analysis

2.1. Plutonium and Uranium Analysis Algorithm

The plutonium and uranium mass determinations are based on a three-factor formulation involving the Pu/U ratio, a correlation relating Pu/U with the quantity (U+Pu)/Uo, and the fabricators specification of the initial uranium mass Uo:

$$Pu = \left[\frac{Pu/U}{1+Pu/U}\right]\left[\frac{U+Pu}{Uo}\right] Uo \qquad (1)$$

The Pu/U ratio is either the value measured by the reprocessor or a value derived from isotope correlations. The two principle ICT-derived Pu/U ratios employed in these initial analyses are: one, from the measured U-235 weight percent using correlation

#1, Dep (U-235) (w/o) vs. Pu/U (g/kg);

and two, from the measured Pu-239 weight percent using correlation

#2, (100 - Pu-239) (w/o) vs. Pu/U (g/kg).

The algorithm produces both the plutonium and uranium masses that "are actually" in the tank if the Pu/U ratio is measured by the reprocessor or inspector and the masses that "should-be" in the tank if the Pu/U ratio is derived from correlations. The first factor identifies the mass value to be determined; that is,

a. the mass that "actually is" in the tank for the measured Pu/U ratio, and
b. the mass that "should-be" in the tank for the ICT-derived Pu/U ratio.

The second factor is computed from the correlation relating (U+Pu)/Uo to the Pu/U ratio used in the first factor. The third factor is the fabricator's specification of the total initial uranium content for the assemblies in the batch. The equation for the uranium mass is analogous, with a 1 replacing the Pu/U in the numerator of the first factor.

2.2 Intermittent Irradiation

The analysis identified anomalies in the irradiation histories of many assemblies; with some assemblies being irradiated for one or two cycles, withdrawn from the configuration for as many as three cycles, and subsequently reinserted for additional burn cycles. First-order correction factors, independent of specific fuel cycle parameters, were computed to correct the isotopics and Pu/U ratio for this effect. The correction factors ranged from 0.3% to 3.0% for the Pu/U ratio. The correction improves the agreement between the measurement-derived and ICT-derived mass determinations for the entire campaign and reduces the overall uncertainty. The correction factors are dependent only upon the number of cycles for which the assembly was removed and not upon the specific fuel cycle numbers. These generic corrections are incorporated into the diagnostic system.

2.3 Single-Correlation Analysis

The analysis of the ESARDA Set A and Set B data using the single correlation scheme are presented both with and without corrections for intermittent irradiation. Correlation #1 and #2 used for the analysis are the calculated theoretical enrichment-dependent correlations modified by the ESARDA Set A data with intermittent irradiation corrections included.

2.3.1 Discussion of Set A Analysis: A typical summary table output for the single correlation analysis is presented in Table I for correlation #1 with intermittent irradiation corrections. The campaign averaged percent difference in the plutonium masses determined from the measurement-derived Pu/U ratio and from the ICT-derived Pu/U ratio obtained from the measured U-235 isotopics as listed in the table is (-0.02 +/- 1.14)% for correlation #1 with intermittent irradiation correction included. If the corrections were omitted, the difference in plutonium mass determination would be (0.84 +/- 1.59)% for correlation #1. The effect of intermittent irradiation corrections are shown in Figure 1 for the 3.10% enrichment batches of Set A, where open diamonds represent the data without correction and the crosses, the data with correction. The correction effected a reduction in the spread of the reprocessing data and deviation from the reference correlation curve. The results for correlations #1 and #2, with (w) and without (w/o) intermittent irradiation (II) corrections on plutonium mass determination are summarized:

Correlation	w/o(II)	w(II)
#1	(0.84 ± 1.59)%	(-0.02 +/- 1.14)%
#2	(0.82 ± 1.74)%	(0.50 +/- 1.54)%

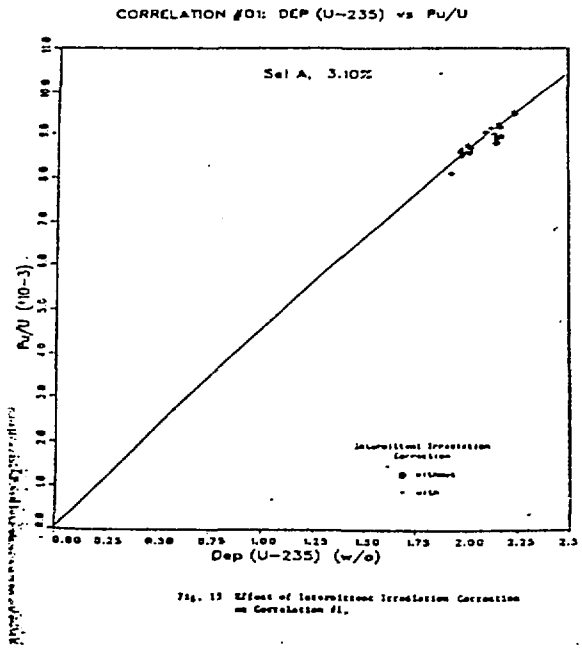

Fig. 1 Effect of Intermittent Irradiation Correction on Correlation #1.

2.3.2 Discussion of Set B Analysis: The single correlation analysis of Set B data with the intentional anomalies included and for correlations #1 and #2 with intermittent irradiation corrections are summarized below. The campaign average percent differences in the plutonium mass determined from the measurement-derived and ICT-derived Pu/U ratios are:

(2.30 +/- 1.86)% for correlation #1
(2.62 +/- 1.46)% for correlation #2

A pronounced increase in the percent difference between the measurement-derived and ICT-derived plutonium mass determination was observed for both correlation between Set A and Set B data. These differences reflect an apparent bias in the Pu/U ratio of about 2-3% between the batches in Set A and Set B. The source of this bias was determined to be the introduction of mixed-oxide fuel assemblies in the loading configurations. Set A assemblies, mainly irradiated in later power cycles with significant numbers of MOX assemblies; Set B assemblies were irradiated in earlier power cycles having few, if any, MOX assemblies. Scoping calculations verified that the presence of MOX fuel assemblies would produce a bias qualitatively consistent with the observed discrepancy between Set A and Set B. Consequently the

analysis is to be performed with isotopic correlations adjusted for the MOX loading configuration.

2.3.3 U-236 Initial Concentration: For both Set A and Set B of the reprocessing data, the correlation between U-235 and U-236 reveals a definite systematic underprediction of the U-236 weight percent, as identified in the U-235 vs. U-236 correlation. For the 3.10% enrichment batches in Set A, the magnitude of this bias is 0.0172 +/- 0.0011 w/o; for the 3.10% enrichment batches in Set B, the bias is 0.0186 +/- 0.0009 w/o. The isotopic correlation analysis identifies that the source of this bias is an initial concentration of U-236 in the uranium fuels. Analysis of the compositions of the uranium fuels used in the ZPR critical facilities at ANL corroborates this concentration level. The U-235 and U-236 weight percents for six uranium oxide or metallic fuels, spanning nearly the complete range of enrichments, represent a dependence of U-236 concentration on uranium enrichment. Interpolation of this fitted curve yields a U-236 concentration of 0.0272 w/o at 3.10% enrichment. The value is in agreement with the U-236 bias in the reprocessing data.

3. ESARDA Diagnostic Analysis

3.1 Diagnostic ICT Analysis

Two aspects of the ANL/ICT diagnostics methodology are (1) to analyze anomalies in the dynamics of isotope production through detail reactor physics computations and (2) to evaluate internal consistency in batch reprocessing data and to identify and quantify any inconsistency. This use of reactor physics computations as diagnostic tools resolved the apparent anomalies in the reprocessing data of the ESARDA exercise. The apparent anomalies were correctly modeled in the deviation and application of the correlations. This feature could be employed after-the-fact to diagnose apparent anomalies discovered in reprocessing data or before-the-fact to predict the sensitivity of ICT to potential anomalies. Results from predictive computations indicate additional safeguards objectives that can be achieved with isotope correlations.

The single-correlation method of mass determination based on a limited number of primary correlations, such as correlations #1 and #2, does not evaluate the consistency among the isotopic and Pu/U reprocessing data or to estimate the plutonium and uranium masses based on the complete set of measured isotopics. A diagnostic analysis algorithm has been developed to incorporate these capabilities in the ANL/ICT method.

Based upon isotopic correlations derived from detailed reactor calculations and adjusted with reliable historic data, this algorithm examines seven measured quantities: U-235, U-236, Pu-239, Pu-240, Pu-241, Pu-242, and Pu/U, each in relation to the other six. The results of these forty-two comparisons are evaluated to determine inconsistencies among the seven quantities and to provide a single, unified ICT-derived Pu/U ratio for mass determinations via the three-factor formula.

Statistical parameters are establish for each correlation characteristic deviations which are to be expected from consistent batch reprocessing data and therefore deviation thresholds which, if significantly exceeded, indicate potential anomalous data. Two statistical parameters are required for the diagnostic implementation: the mean and standard deviation for the distribution of deviations of the reference data from each adjusted correlations, both quantities being expressed in weight percent for the isotopics and in gm/kg for the Pu/U ratio.

3.2 Diagnostic Algorithm

The algorithms have been developed to evaluate in a unified manner the measured data against the complete set of diagnostic correlations. One aspect of this evaluation is to investigate the internal consistency of the reprocessing data, that is, to examine the isotopics and Pu/U ratio applying all of the correlations in order to detect systematic deviations which portend anomalies in the data. A second aspect is to utilize all of the measured isotopics in order to ascertain the best estimate of the plutonium and uranium masses. This approach decreases the overall uncertainty in the estimated masses and reduces the potential for biases introduced by mass determinations reliant on a single isotopic weight percent.

The diagnostic algorithm is structured to compute the deviations of the reprocessing data from the set of adjusted correlations, to determine batch enrichment anomalies, to diagnose isotopic and Pu/U ratio outliers, and to determine the plutonium and uranium masses for the measured and ICT-derived Pu/U ratios.

4. ESARDA Diagnostic Analysis Results

4.1 Discussion of Intentional Anomalies

The initial application of diagnostic routine was the analysis of the ESARDA Set B reprocessing data. Eleven of the twenty-nine batches in this data set had been intentionally modified to provide a means to

compare the diagnostic capabilities of various ICT methods in the reprocessing community. Referring to Table II, there were three categories of anomalies introduced:

1. misstated assembly burnup, (batches: 14, 26)

2. misstated assembly content of batch, and (batches: 3, 8, 21, 29)

3. misstated isotopics and/or Pu/U ratio. (batches: 6, 12, 14, 17, 18,27)

The misstatement of the burnup occurred either alone (batch 26) or in conjunction with a misstatement of the Pu/U ratio (batch 14). The misstatement of the assemblies constituting the batch manifested itself through erroneous values in other quantities, such as, initial enrichment, effective burnup, decay correction factors, and intermittent irradiation factors.

4.2 Diagnostics of Intentional Anomalies

Table II, summarizes the intentional anomalies for the eleven modified batches and also identifies and quantifies the anomalies detected by the diagnostic analysis scheme. Anomalies in U-234 and Pu-238 were not deduced since these isotopics were not included among the indices in the diagnostic routine. The only anomaly not resolved was a 0.4 w/o modification to Pu-239, with a nominal concentration of 60 w/o, in batch 12. Since the ANL method does not utilize assembly burnup in any phase of its isotopic correlation analysis, the diagnostics scheme does not recognize a misstatement of burnup as anomaly; that is, the method diagnoses anomalies in the measured quantities without requiring an accurate estimate of the calculated burnup, as shown in baten 14.

The rightmost column in the Table II, signifies the difference in the deduced anomaly from the intended value in units of standard deviation of the uncertainty in the deduced anomaly. The magnitude of the deduced anomaly considers the deviation of the modified data from the correlation; the statistical error in the data before the anomaly was introduced is a component of this difference. Errors associated with the diagnostic scheme yield another component. Since statistical uncertainties for the reprocessing data were not specified, the contributions to the difference from these sources cannot be estimated.

Referring to Table III, large percent differences (-3½ %) exist between the measurement-derived and ICT-derived Pu/U ratio and mass determinations for batches B14 and B27. The reason for this is that the measured data contained large intentional anomalies in the Pu/U ratio which were detected by the diagnostic analysis; the diagnostics computed estimates of the mass content based on ICT-derived Pu/U ratios consistent with non-suspect isotopics and hence without the intentional anomalies.

4.3

4.4 Diagnostic Methodology

The ANL diagnostic methodology provides an ICT-derived mass content that "should-be" in the tank and identifies and quantifies anomalies which may result from the intentional falsification of reprocessing data, measurement errors and biases, off-normal power reactor and/or reprocessing plant operations, and fabricators initial uranium specifications.

5. Summary

The isotopic correlation technique analysis and diagnostic algorithms have been developed at Argonne National Laboratory for implementation on mainframe and personal computer systems. The analysis and diagnostic software has the capability to incorporate facility-dependent features and can be implemented for infield and/or onsite inspectorate activities. The implementation of the ANL/ICT system for the ESARDA exercise has demonstrated that ICT can be used as an independent verification for determining the mass input to the reprocessing plant and as an instrument for diagnosing anomalies in the fuel cycle from fabrication, power reactor and reprocessing plant operations, and in measured reprocessing data. The aspects of the ESARDA study were:

- the identification of abnormal power reactor operating procedures, the intermittent irradiation of assemblies, which yielded apparent anomalous data and the effect of physics-based correction factors;

- the discovery of an unintentional bias of 2-1/4% between the single correlation analysis of Set A, the reference data and the identical analysis of Set B, the test data;

- the resolution of the source of an apparent bias, using ICT diagnostic reactor physics computations, as being the presence of partial mixed-oxide loadings

during the irradiation of the reprocessed assemblies;

- the successful detection of the anomalies intentionally introduced into the Set B reprocessing data to evaluate ICT diagnostic capabilities; and

- the improvement, using the diagnostic analysis, in the agreement between the ICT-derived and measurement-derived mass determinations for Set B on the batch basis.

6. References

1. Christensen, D.E., Schneider, R.A. "Summary of Experience with Heavy-Element Isotopic Correlations." Safeguarding Nuclear Materials, Proc. IAEA Symp., Vienna 1975, Vol. II, pp. 277-393. 1975.

2. Bouchard, J., Dean, G., Patigny, P., Robin, M. Possibilities Offered by the Analysis of Fuel History for Safeguards Surveillance at the Input End of Reprocessing Plants. Safeguards Technology - 1978, Proc. IAEA Symp., Vienna 1978, Vol. II, pp. 777-795 (in French). 1978.

3. Timmerman, C.L., Selby, G.P., and Napier, B.A., "Selected Isotopic Functions: A Description and Demonstration of Their Uses," ISPO-37, PNL-2761 (October 1979).

4. L. Koch and S. Schoof, "The Isotope Correlation Experiment," ESARDA Final Report ESARDA 2/81, EUR7766EN, KfK3337, (July 1982) Published by Commission of the European Communities Joint Research Centre, ISPRA, Italy,

5. "Gravimetric Method (Pu/U Ratio) for Input Measurements," Tokai Advanced Safeguards Technology Exercise (TASTEX), Report No. 213, pp. 187-191, IAEA, Vienna (1982).

6. Aries, M., Patigny, P., Bouchard, J., Giacometti, A., Girieud, R. "Quatre Ans d'Experience d'Utilisation des Correlations Isotopiques Calculees dans l'Etablissement du Bilan d'Entree a l'Usine de la Hague." Nuclear Safeguards Technology 1982, IAEA Symp., Vienna, (November 1982), Vol. I., pp. 455-471. 1983.

7. Giacometti, A., Girieud, R., Aries, M. "Calculated ICT Improvements: Consequences of Five Years of Industrial Implementation." ESARDA, 6th Annual Symp. on Safeguards and Nuclear Material Management. May 1984.

8. Giacometti, A. "ESARDA Exercise to Test performance of ICT Procedure." Proc. ESARDA, Sixth Ann. Symp. on Safeguards and Nuclear Materials Management, Venice, Italy, 14-18 May 1984, pp. 213-222. May 1984.

9. Bouchard, J., Patigny, P. "Techniques for Material Control at the Input of a Reprocessing Plant." Proceedings of the Conference on Safeguards Technology: The Process - Safeguards Interface, Hilton Head Island, SC, 28 November - 2 December 1983, CONF-831106, pp. 348-355. August 1984.

10. P.J. Persiani, Kalimullah, R.G. Bucher, and R.B. Pond, "Isotope Correlations for Safeguards and Accountability Methods for Light Water Reactor Fuel Cycles, Proceedings of Conference on Safeguards Technology: The Process-Safeguards Interface, November 28 - December 2, 1983, (ANS/INMM) Hilton Head Island, South Carolina Conf.-831106, pp. 204-216, August 1984.

Table I. Mass Determination and Pu/U Ratio by ANL/ICT Method
COGEMA Reprocessing Input Mass Verification
Obrigheim PWR Spent Fuel Assemblies
Set A Data, at Reprocessing
with Intermittent Irradiation Corrections
Correlation #1

Batch Label	Pu/U Ratio (gm/kg) Meas Pu/U	ICT Pu/U	DIFF (%)	Pu Mass (gm) Meas Pu/U	ICT Pu/U	DIFF (%)
A 01	7.933	8.175	3.05	4237.9	4361.1	2.91
A 02	8.517	8.509	-.10	4355.2	4351.1	-.09
A 03	8.273	8.297	.29	4408.7	4420.9	.28
A 04	8.483	8.534	.60	4506.6	4532.3	.57
A 05	8.475	8.367	-1.28	4470.7	4416.5	-1.21
A 06	8.125	7.953	-2.12	4148.1	4064.4	-2.02
A 07	8.802	8.794	-.09	4681.4	4677.3	-.09
A 08	9.220	9.189	-.34	4891.2	4875.6	-.32
A 09	9.329	9.281	-.51	4928.9	4905.1	-.48
A 10	9.003	8.998	-.06	4771.0	4768.5	-.05
A 11	9.075	9.057	-.19	4868.9	4860.0	-.18
A 12	8.647	8.796	1.73	4575.5	4650.1	1.63
A 13	8.489	8.669	2.13	4500.7	4590.9	2.00
A 14	8.798	8.871	.83	4651.7	4688.3	.79
A 15	8.754	8.890	1.56	4646.0	4714.2	1.47
A 16	8.411	8.290	-1.44	4463.1	4401.9	-1.37
A 17	8.363	8.270	-1.11	4453.9	4406.9	-1.05
A 18	8.422	8.362	-.71	4457.8	4427.9	-.67
A 19	8.476	8.510	.40	4514.4	4531.7	.38
A 20	8.547	8.432	-1.35	4570.4	4512.0	-1.28
A 21	8.883	8.823	-.68	4753.6	4723.2	-.64
A 22	8.973	8.915	-.65	4799.0	4769.5	-.61
A 23	8.817	8.857	.45	4642.6	4662.7	.43
A 24	8.415	8.341	-.87	4466.3	4429.2	-.83
Campaign Average = & Uncertainty			-.02 +/- 1.20			-.02 +/- 1.14

Table II. Evaluation of Diagnostic Analysis Results for Obrigheim Set B Data

Batch Label	Quantity	COGEMA Intentional Anomaly	ANL ICT Diagnosed Anomaly	COGEMA-ANL Difference (Std Dev)
B 03	Enrichment	-0.131	-0.149 +/-0.034	0.5
B 06	U-234	-0.002	(a)	(a)
	U-235	0.057	0.065 +/- 0.010	0.8
	U-236	0.011	0.012 +/- 0.001	1.0
	U-238	-0.066	-0.077 +/- 0.010	1.1
B 08	Enrichment	0.135	0.120 +/- 0.028	0.5
B 12	Pu-239	0.400	(b)	(b)
	Pu-242	-0.400	-0.376 +/- 0.039	0.6
B 14	Pu/U	-0.300	-0.280 +/- 0.023	0.9
	Burnup	(Burnup not used in ANL ICT		)
B 17	U-235	0.050	0.068 +/- 0.005	3.6
	U-238	-0.050	-0.068 +/- 0.005	3.6
B 18	Pu-238	-0.002	(a)	(a)
	Pu-239	0.840	0.972 +/- 0.140	0.9
	Pu-240	-0.753	-0.750 +/- 0.048	0.1
	Pu-241	0.237	0.153 +/- 0.117	0.7
	Pu-242	-0.322	-0.414 +/- 0.040	2.3
B 21	Enrichment	-0.042	-0.048 +/- 0.028	0.2
B 26	Burnup	(Burnup not used in ANL ICT		)
B 27	Pu/U	-0.259	-0.328 +/- 0.067	1.0
	Pu-238	-0.025	(a)	(a)
	Pu-240	-0.722	-0.875 +/- 0.133	1.1
	Pu-241	0.747	0.811 +/- 0.089	0.7
B 29	Enrichment	0.056	0.052 +/- 0.026	0.2

(a) Anomalies in U-234 and Pu-238 not included in diagnostic algorithm.

(b) Anomaly undetected by diagnostics.

Table III. Mass Determination and Pu/U Ratio by ANL/ICT Method
COGEMA Reprocessing Input Mass Verification
Obrigheim PWR Spent Fuel Assemblies
Set B Data, at Reprocessing
Diagnostic Analysis

Batch Label	Pu/U Ratio (gm/kg) Meas Pu/U	ICT Pu/U	DIFF (%)	Pu Mass (gm) Meas Pu/U	ICT Pu/U	DIFF (%)
B 01	8.095	8.216	1.49	4531.6	4595.6	1.41
B 02	8.147	8.235	1.08	2281.3	2304.7	1.03
B 03	8.288	8.274	-.17	4511.1	4503.8	-.16
B 04	8.211	8.273	.76	4597.0	4630.0	.72
B 05	8.256	8.243	-.15	4515.9	4509.4	-.15
B 06	8.304	8.369	.78	4439.5	4472.2	.74
B 07	8.249	8.211	-.46	4372.1	4353.2	-.43
B 08	8.385	8.396	.13	4455.6	4461.1	.12
B 09	8.605	8.716	1.29	4533.6	4588.5	1.21
B 10	8.140	8.142	.02	4367.7	4368.5	.02
B 11	8.122	8.154	.40	4326.5	4342.9	.38
B 12	8.178	8.140	-.46	4341.0	4322.0	-.44
B 13	8.305	8.413	.131	4427.3	4481.9	1.23
B 14	8.672	8.392	-3.23	3465.4	3359.7	-3.05
B 15	8.180	8.168	-.15	5474.9	5467.1	-.14
B 16	8.211	8.336	1.52	4374.4	4437.5	1.44
B 17	8.223	8.229	.07	4404.0	4407.1	.07
B 18	8.237	8.152	-1.03	6544.5	6480.4	-.98
B 19	8.129	8.136	.09	6476.3	6481.7	.08
B 20	8.223	8.201	-.27	6562.5	6546.0	-.25
B 21	8.258	8.220	-.46	4429.8	4410.5	-.44
B 22	8.066	8.033	-.41	5394.5	5373.4	-.39
B 23	8.307	8.377	.84	5535.3	5579.2	.79
B 24	8.785	8.660	-1.42	5812.0	5734.1	-1.34
B 25	8.588	8.439	-1.74	5691.3	5597.8	-1.64
B 26	8.363	8.284	-.94	5569.2	5519.5	-.89
B 27	8.569	8.241	-3.83	5698.3	5491.4	-3.63
B 28	8.403	8.342	-.72	5589.5	5551.2	-.69
B 29	8.554	8.504	-.59	3413.1	3394.3	-.55
Campaign Average = & Uncertainty			-.22 +/-1.23			-.20 +/- 1.16